# Photoreflectance, absorption, and nuclear resonance reaction studies of $Al_xGa_{1-x}As$ grown by molecular-beam epitaxy

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The photoreflectance (PR) spectra of bulk $Al_xGa_{1-x}As$ alloys with $x \leqslant 0.45$ were studied. The observed line shapes from different samples suggest that the PR technique is very sensitive to the material quality, surface condition, and the background impurities. The energy gap derived from the PR spectra compared well to that obtained from the absorption spectra. The relationship between the energy gap and the Al mole fraction value $x$ was established through the nuclear resonance reaction analysis. The electric field near the surface was calculated from the periodicity of Franz–Keldysh oscillations observed in many of the samples. From our analysis, we believe that the number of oscillations shown in PR spectra corresponds to sample quality, in general. We also believe that the low-field-like line shape is mainly caused by the fluctuation of Al distribution along the growth direction. An additional feature related to the impurity transition was also observed in the spectra.

## I. INTRODUCTION

As one of the reflectance modulation techniques, photoreflectance (PR) experiments were first reported about two decades ago.[1] The fundamentals of PR spectra obtained in semiconductor material such as GaAs were also reported early on.[2–5] Most recently, however, PR has received a great deal of attention because it is well suited for studying novel materials grown by, e.g., molecular-beam epitaxy (MBE). The materials studied include GaAs/AlGaAs superlattices,[6] modulation-doped heterojunctions,[7] GaAs quantum wells grown on Si,[8] and the InGaAs/GaAs strained layer superlattices.[9] Allowed, forbidden, as well as the so-called unconfined transitions in GaAs/AlGaAs quantum well structures (QWSs) have been investigated.[10,11]

The advantages of PR over other techniques are the simplicity of its measurement setup, its nondestructive nature, and the sharp line shapes that can be observed even at room temperature. In the study of semiconductors, it is believed that the origin of PR is similar to that of electroreflectance. The modulation is due to the change in the electric field at the Schottky surface barrier by photoexcited carriers.[3] The electroreflectance theory introduced by Aspnes[12] has been used to explain the PR results on semiconductor superlattices. Despite apparent success, a detailed framework remains to be established about the nature of PR line shapes, as indicated by Shanabrook *et al.*[13] As in QWSs, the PR spectra of bulk materials such as GaAs and $Al_xGa_{1-x}As$ seem also complicated and difficult to analyze. The reason is not only that PR is a passive method but the surface band bending cannot be controlled as well as that in electroreflectance. Also, the transitions, such as the heavy hole, light hole, and those associated with impurities, are usually mixed together.

We chose to investigate the $Al_xGa_{1-x}As$ alloy because it is an important alloy used for many devices. Its optical properties, especially the photoluminescence and absorption, have been reasonably well understood. By comparing PR spectra with the results from other measurements, one can gain more insight about the nature of PR in $Al_xGa_{1-x}As$. From the practical point of view, $Al_xGa_{1-x}As$ is a widely used alloy system in semiconductor heterojunctions, both in optical and electronic devices. Although there are many techniques which have been used to characterize the material properties, especially the AlAs mole fractions and the sample quality, the limitations are obvious. Ordinary x-ray analysis, which is often employed to measure the lattice constant, suffers from the large penetration depth and lacks the depth resolution. Photoluminescence gives a sharp signal at the energy related to the energy gap at low temperatures, but the signal is broadened and the intrinsic transition is usually mixed with impurity-related ones at room temperature. Thus, a relatively large uncertainty is introduced in the determination of the energy gap of the alloy. From the low-temperature absorption spectrum, the excitonic transitions signal can be observed for $x$ values as large as 0.4.[14] However, in addition to the requirement of low temperatures, transmission measurement is useful only from a research point of view since the GaAs substrate must be removed in order to observe the transition with energies larger than the GaAs fundamental gap. A new method, nuclear resonant reaction analysis (NRRA),[15–17] for the direct and nondestructive determination of the Al concentration in GaAs/AlGaAs heterostructures can be employed. This measurement provides a very sensitive and accurate tool for Al depth profiling with depth resolution of a few hundred angstroms but needs sophisticated and expensive equipment, such as an accelerator. Compared with the techniques mentioned above, PR is becoming a promising and an extremely valuable tool because of the advantages stated above. The remaining problem is how to relate the measured spectra to the material parameters and characteristics.

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In this paper, systematic studies of PR results on $Al_xGa_{1-x}As$ with the AlAs mole fraction varying from 0 to 0.45 near the fundamental band gap are presented. We have observed different line shapes from various samples, which suggests that PR is very sensitive to the nonuniformity of alloy composition, the surface condition, and the background impurities. The transition energies of the fundamental gap were derived and compared with those obtained from the low-temperature absorption measurements. The agreement between the PR and the absorption results demonstrates that PR can be used as a highly accurate and convenient technique. We also report the observation of the PR spectrum related to the impurity transitions. The estimates of the broadening parameters through the linewidth and the surface field through the period of the Franz–Keldysh oscillations make it possible to get information about the alloy quality in the bulk and at the surface (or interface).

In addition to the optical measurements, Al depth profiling by NRRA[15–17] has been used to determine the AlAs mole fraction. Thus, the relationship between the energy gap obtained from optical measurements and the $x$ value from NRRA method is established. The results are compared with the previously reported data.[18]

## II. EXPERIMENTAL PROCEDURES

The samples used in this study were grown by MBE on (100) GaAs substrate. The typical growth sequence consisted of a 0.2-$\mu$m GaAs buffer layer followed by the ternary alloy. The $Al_xGa_{1-x}As$ epilayer thicknesses ranged from 1 to 3 $\mu$m and most were nominally undoped. The $p$-type background impurities were estimated to be in the $10^{15}$ $cm^{-3}$ range at 300 K and may increase with $x$. A very thin ($\sim$300 Å) AlAs etch stop layer was introduced between the buffer and epilayer for alloys with low Al compositions to facilitate the preparation of samples for optical transmission measurements.

The details of transmission experiments can be found in Ref. 19. Since the absorption edge of the GaAs substrate and buffer layers is lower than the $Al_xGa_{1-x}As$ epilayer, the GaAs must be removed. The steps include mechanical lapping and chemical polishing, and finally selective chemical etching of a 1-mm-diam photolithographically defined window. Measurements were performed with samples cooled to about 4 K in a Janis optical cryostat so that the absorption due to the sharp excitonic resonance can be observed.

The NRRA technique has been used extensively to provide nondestructive, direct, and precise analysis of elemental concentrations in a sample and accurate depth profiling with a high resolution below the surface. Its principle has been well described elsewhere.[15–17] Its use for element analysis and depth profiling depends on the existence of an isolated $\delta$-functionlike resonance in the nuclear reaction involving the appropriate isotope to be analyzed. Al depth profiling makes use of the nuclear reaction[27] Al $(p,\gamma)$ $^{28}$Si which has a very narrow isolated resonance at a proton energy, $E_{lab}$, of 992 keV with a resonance width $\Gamma$ of about 100 eV. In the measurement the sample is bombarded with protons in an appropriate energy range higher than the resonance energy 992 keV, and the characteristic $\gamma$ rays of 10.7 MeV emitted promptly during de-excitation of the excited residual nucleus $^{28}$Si in a reaction are detected. The $\gamma$-ray yield per incident ion is proportional to the amount of Al at the depth where the reactions occur. Selection of the incident proton energy is equivalent to specifying the depth at which the Al concentration is to be determined since the incident ion loses part of its kinetic energy down to the required resonant energy when it penetrates a certain depth in the target. Thus, measuring $\gamma$-ray production as the incident beam energy is varied gives a direct measurement of Al concentration as a function of depth in the target.

Our analysis is conducted with the Caltech EN-Tandem accelerator. The experimental setup was the same as described in Ref. 15, which is similar to those for hydrogen depth profiling presented in Ref. 16. The targets were mounted in a special chamber at the end of the beam line maintained under a vacuum of $10^{-6}$ Torr. The beam was focused and directed through a 2-mm-diam collimator onto the target with a current about 30 nA. A $20\times15$ cm NaI (Tl) scintillation detector, placed at 90° to the incident beam direction and about 3 cm from the target, was used for $\gamma$-ray detection. During the measurement the $\gamma$-ray yield was taken from an energy window of the $\gamma$-ray spectrum. Conversion of the incident beam energy to the depth scale of the target was made with using the the stopping cross section, $dE/dx$, which can be found in standard references.[20] The interpretation of the yield data as a function of the incident beam energy in terms of the Al concentration versus depth was made with aid of a calibration constant which was obtained from a measurement of an AlAs sample, after correcting for the energy-loss difference of incident beam in the various target materials.

The experimental arrangement for photoreflectance is similar to that discussed in Ref. 3. A white light from a 100-W tungsten-halogen source passes through a model 270 McPherson monochromator to produce the monochromatic radiation that probes the crystal surface. The reflected light was detected by a EG&G type HUV-1100 (BG) Si photodiode using the usual lock-in detection technique. For the samples with small $x$ values, a 1-mW He-Ne laser (6328 Å) was used as a pump beam and mechanically chopped at 90 Hz to modulate the band structure of the crystal surface. For the samples with $x\leqslant0.35$, the krypton ion laser (5682 Å) was used. Several neutral density filters were used to reduce the laser power to about 100 mW/cm$^2$. The output of the lock-in amplifier was recorded by an $X$-$Y$ recorder.

## III. RESULTS AND DISCUSSION

### A. Absorption

The absorption spectrum of pure bulk GaAs is dominated by excitonic peaks at low temperatures. For the $Al_xGa_{1-x}As$ alloys, similar spectra can be observed if the AlAs mole fraction is in the direct transition region ($0\leqslant x<0.50$). For samples with higher $x$ values, the excitonic resonance is usually less strong and broader than that of lower $x$ values. The resonant strength and broadening also depend on the quality of the crystals. Figure 1 gives one of the transmission spectra obtained directly from our mea-

surement for the sample with $x = 0.30$. The good quality of this epilayer is evident from the strong excitonic structures dominating the spectrum. Two absorption peaks indicated by the arrows in Fig. 1 are attributed to the transitions corresponding to $n = 1$ and excited states of free excitons.[14] Since the energy of these excitonic transitions in transmission spectra can be accurately determined, the fundamental band gap $E_g$ can be derived precisely from the equation

$$E_g = E_1 + E_b, \tag{1}$$

where $E_1$ is the observed transition energy of $n = 1$ state of free exciton, and $E_b$ is the exciton binding energy which is obtained from the energy difference between the $n = 1$ and $n = 2$ (or first excited) states of excitonic transitions[14]:

$$E_b = 1.35(E_2 - E_1). \tag{2}$$

### B. Depth profiling by NRRA

NRRA technique was used in more than 10 $Al_xGa_{1-x}As$ samples with different AlAs mole fractions. Shown in Fig. 2 is the detected photon counts of the $\gamma$-ray yield as a function of incident proton energy for some samples. For proton energies lower than 992 keV, a very small number of protons was detected since the energy is too low for the nuclear reaction to occur even at the sample surface. For proton energies higher than 992 keV, the protons can penetrate a certain average distance before their energies are reduced to 992 keV, and resonant reaction between the protons and $^{27}Al$ can occur. The ray yield is proportional to the Al concentration at that distance from the surface of the material. The Al signal was compared to that obtained in the AlAs sample for conversion to mole fractions.

From Fig. 2, one can see clearly the depth profiling of $^{27}Al$. Samples 1583, 1982, and 1816 show a more uniform distribution of Al atoms along the growth direction than the others. The best quality of sample 1583 observed in NRRA

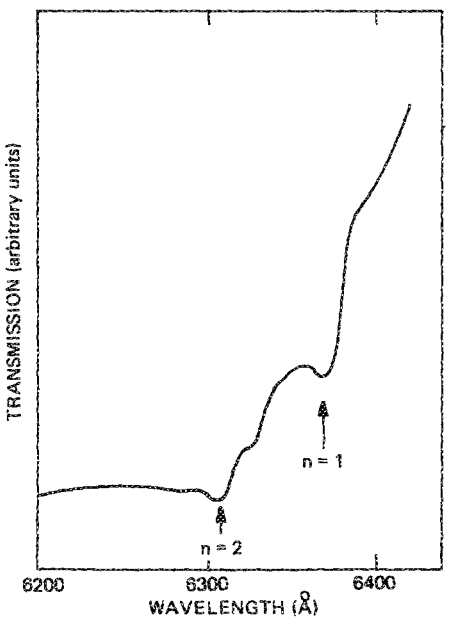


FIG. 1. Transmission spectrum from sample 1583 with $x = 0.30$. Arrows indicate the excitonic resonances of $n = 1$ and first excited states.

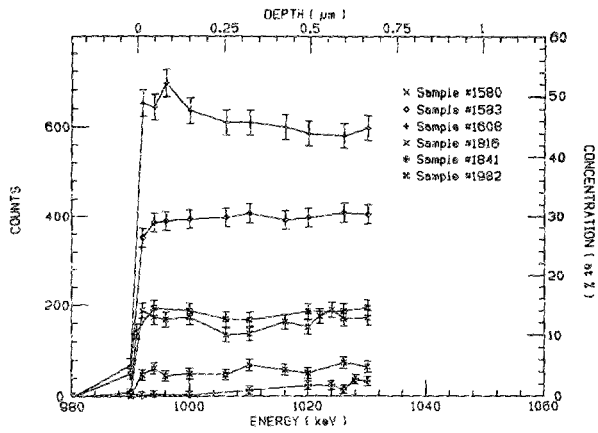


FIG. 2. Al concentration profiling determined from NRRA for several samples.

was also proven by the absorption, photoluminescence, as well as the PR spectra which will be discussed later. The absolute measurement error in AlAs mole fraction $x$ is estimated to be 0.006 for sample 1583, corresponding to a relative deviation of about 2%. One can see that for good quality (uniform Al distribution along the growth direction) samples, the uncertainty in the AlAs mole fraction is dominated by the intrinsic errors of the measurement technique. For other samples, the uncertainty of the AlAs mole fraction is determined by the inhomogeneous distribution of the Al in alloy, which manifests itself as broadening of the observed optical features.

### C. Photoreflectance

The PR spectra near the fundamental band gap of $Al_xGa_{1-x}As$ at room temperature are more complicated than those of GaAs/AlGaAs quantum wells and even bulk GaAs. Different line shapes were observed in samples with different $x$ values. In Figs. 3 and 4, we show two typical PR spectra with one having the line shape similar to that predicted by Aspnes third derivative functional form (TDFF)[12] and the other showing clearly the Franz–Keldysh oscillations.

The spectrum shown in Fig. 3 was taken from sample 1841 with $x = 0.02$. The spectrum has fewer oscillations and

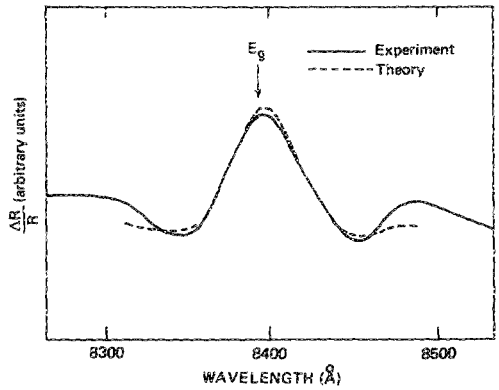


FIG. 3. PR spectrum from sample 1841 with $x = 0.02$. The broken line is the theoretical fit to Eq. (3).

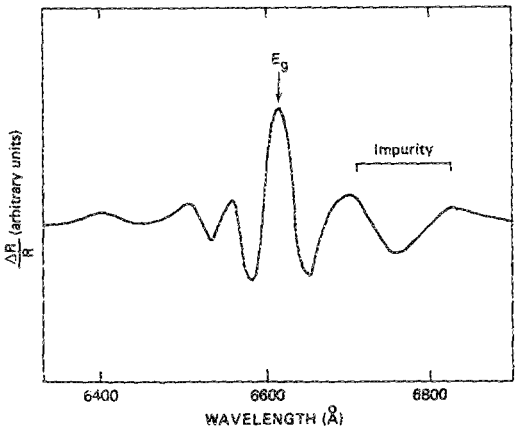


FIG. 4. PR spectrum from sample 1583 with $x = 0.30$, which shows Franz–Keldysh oscillations as well as the impurity-related structure.

should fall into low-field modulation domain (either low field or large broadening) and can be fitted to Aspnes TDFF[12]:

$$\Delta R/R = ce^{i\Theta}(E - E_g + \Gamma)^{-m}, \tag{3}$$

where $c$, $\Theta$, $E_g$, and $\Gamma$ are the amplitude, phase, transition energy at a critical point, and broadening parameter, respectively. Here, $m = 2.5$ for the three-dimensional $M_0$-type critical point investigated in the present work. The energy gap and the broadening parameters are determined directly by fitting the experimental line shape to Eq. (3). The calculated line shape is sketched by the broken line and a good agreement is obtained. It should be noted that $E_g$ obtained by this method is close to the dominated peak of PR and $\Gamma$ is always very nearly equal to the energy separation between the extrema.[12] The PR spectra from several samples with different $x$ values but having similar line shapes were also fitted to Eq. (3). The broadening parameter is smaller than 10 meV for $x \leqslant 0.19$ and usually increase with $x$.

The PR spectrum shown in Fig. 4 was taken from the sample with $x = 0.30$. In order to understand how many transitions are involved in this spectrum, we performed the PR measurements under different pump beam intensities. We found that there exist at least two structures which can be distinguished by different dependence of the line shapes on the modulation intensity. The structure with higher energy is related to the band-to-band transition since no other transition with such a strong signal is possible. The oscillations, known as the Franz–Keldysch oscillations, imply that the modulation does not fall into the low-field domain and it is difficult to fit such a line shape to Eq. (3). However, it has been proven from both the theory[12] and experiment[3] that the critical point should be near the energy of the first (low-energy side) dominant peak in the oscillations in PR spectrum for a $M_0$ critical point. This peak energy should not be very much affected by the intensity of the modulation beam. This is really the case in our observations. Thus, the band gap of the sample can be estimated from the position of the first main peak in the oscillatory line shape, as in Fig. 4. Since the energy difference between the two nearest peaks is only about 8 meV, we expect the uncertainty in $E_g$ to be less than 4 meV.

The energy gap of the $Al_xGa_{1-x}As$, determined from the PR spectra as described above, can be compared with that determined from the absorption measurements. It should be noticed that the absorption experiments were carried out at low temperatures. In order to compare it with the PR results, a correction to the energy gap from low temperature to room temperature is necessary. Since the energy gaps of GaAs and AlAs at room and low temperatures are well determined,[21] the correction for $Al_xGa_{1-x}As$ alloys can be obtained by the linear interpolation:

$$\delta(x) = 0.094 + 0.006x \quad \text{eV}. \tag{4}$$

Table I lists the energy gaps derived from absorption and PR spectra for several samples with different $x$ values. One can see that the agreement is quite good. The energy gap deviations are smaller than 11 meV for all of the samples which leads to an uncertainty in the AlAs mode fraction of less than 1%. We believe that the deviations are mainly introduced from the uncertainty in determining exactly the transition energy in PR specta.

Using the AlAs mode fractions obtained from the nuclear profiling technique as described previously, we have plotted the dependence of the energy gap of $Al_xGa_{1-x}As$ alloy on the AlAs mole fraction $x$. The results are shown in Fig. 5. By the least-squares fit of the second-order polynomial $E_g = A + Bx + Cx^2$ to the experimental data including the GaAs energy gap, one can get the constants $A$, $B$, and C. The result,

$$E_g(x) = 1.424 + 1.427x + 0.041x^2 \quad \text{eV}. \tag{5}$$

(for $x \leqslant 0.45$), is also plotted in Fig. 5. We find that the bowing factor $C$ is much smaller than 0.37 reported previously by Lee *et al.*,[22] but comparable to the theoretical values,[23,24] and the most recently published experiment results by Oelgart *et al.*[25] $E_g(c)$ increasing more rapidly with Al mole fraction than the relationship given by Casey and Panish[26] for small $x$ was also reported recently by Aspnes *et al.*[27] and Kuech *et al.*[28]

In some samples structure in PR spectra at energies smaller than $E_g$ is also observed as shown in Fig. 4. We believe that this structure is related to the impurity transitions for the following reasons: (1) The peak intensities of these impurity related features increases more rapidly than that of $E_g$ with increasing modulating beam intensities. The results are similar to those of impurity-related peaks observed in GaAs using electroreflectance.[29] (2) The energy

TABLE I. Comparison of the energy gap determined from the absorption and PR spectra for some of the samples used in this study.

| | | $E_g^{ab}$ (eV) | | $E_g^{PR}$ (eV) | Deviation |
|---|---|---|---|---|---|
| Sample | $x$ | 4 K | 300 K | 300 K | (mev) |
| 1841 | 0.02 | 1.574 | 1.480 | 1.477 | 3 |
| 1816 | 0.05 | 1.583 | 1.489 | 1.483 | 6 |
| 1580 | 0.12 | 1.700[a] | 1.605 | 1.597 | 8 |
| 1982 | 0.14 | 1.713 | 1.618 | 1.607 | 11 |
| 1583 | 0.30 | 1.976 | 1.880 | 1.873 | 7 |
| 1608 | 0.45 | 2.165 | 2.068 | 2.076 | 8 |

[a] Data from photoluminescence spectrum.

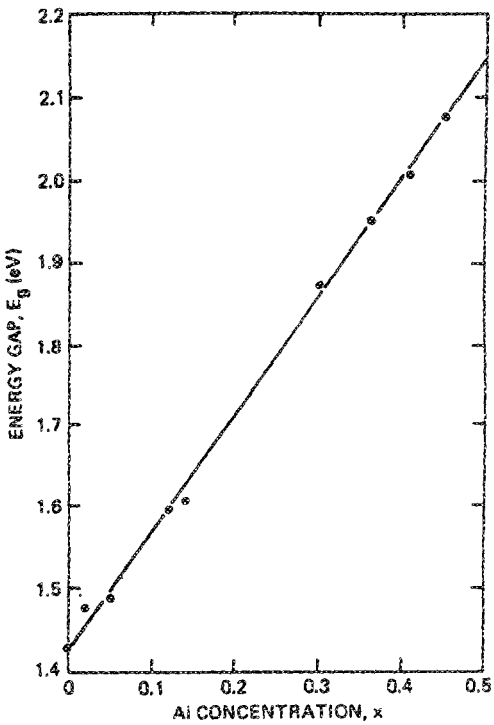


FIG. 5. $Al_xGa_{1-x}As$ band gap as a function of Al concentration determined directly from NRRA. The solid line is the least-squares fit to the experimental data.

difference Δ between these structures and $E_g$ is near the impurity binding energies which can be determined from low-temperature photoluminescence spectra. Figure 6 gives results obtained from PR spectra of several samples. Although the uncertainties are estimated to be about 10 meV due to the difficulties in determining Δ because of the structure mixing, the increase of Δ with $x$ is clear, which is consistent with our PL results and Refs. 30 and 31. We believe that the impurity is $p$ type due to the large binding energy as in the case of unintentionally doped bulk GaAs.[32]

Aspnes[33] determined the interband reduced masses for different critical points in Ge from the Franz–Keldysh oscillations observed in electroreflectance. Using the same method, one can get the strength of the electric field near the $Al_xGa_{1-x}As$ surface if the known reduced mass is assumed. The intermediate field modulation reflectance theory predicts that[33]

$$\Delta R/R \sim C(E)\cos\left[\tfrac{2}{3}(E-E_g/\hbar\Omega)^{3/2}+\theta_0\right] \qquad (6)$$

for $(E-E_g)>\hbar\Omega$, where $C(E)$ is the envelope function of the oscillations that varies slowly with energy on the scale of the argument of cosine term. $\theta_0$ is a constant for a specified critical point. The extrema of the oscillations are determined by

$$\tfrac{2}{3}(E-E_g/\hbar\Omega)^{3/2}+\theta_0=n\pi, \qquad (7)$$

where $n$ is an integer. By plotting $(E-E_g)^{3/2}$ vs $n$, one can obtain a straight line from which the characteristic energy $\hbar\Omega$, in turn the electric field $E_s$, is determined via the slope $S=\frac{3}{2}(\hbar\Omega)^{3/2}$ and where

$$(\hbar\Omega)^3=e^2E_s^2\hbar^2/8\mu_{11}.$$

The straight line shown in Fig. 7 demonstrates that Eq. (7) is also valid for photoreflectance. The characteristic energy $\hbar\Omega$ and the electric field are calculated to be 4.8 meV and 10.3 kV/cm, respectively. Here we used the reduced mass of $Al_xGa_{1-x}As$ by a linear interpolation of the GaAs and AlAs as indicated in Ref. 17, $\mu_{11}=m_hm_e/(m_h+m_e)=0.081m_0$. Since the thoery predicts that the oscillations appear only when the broadening is not much larger than the characteristic energy $\hbar\Omega$, i.e., $\Gamma<3\hbar\Omega$, the broadening parameter can be estimated to be smaller than 14.4 meV.

Several interesting points about the PR spectra from $Al_xGa_{1-x}As$ can be discussed now in some detail. First, two important factors determining PR line shape are the broadening parameter and the surface field, if the inhomogeneous nature of the field is neglected. In electronreflectance, it should be possible to get similar line shapes for different samples by controlling the applied field to yield the same effective parameter $\Gamma/\hbar\Omega$.[34] In PR, however, different line shapes are expected for $Al_xGa_{1-x}As$ samples, as we observed in the experiments, since the surface field is deter-

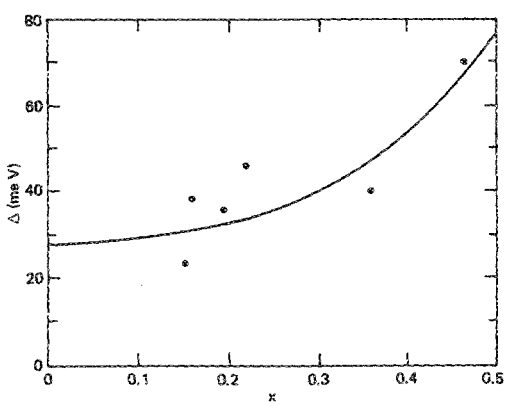


FIG. 6. Impurity binding energies Δ of $Al_xGa_{1-x}As$ estimated from PR spectra as a function of $x$.

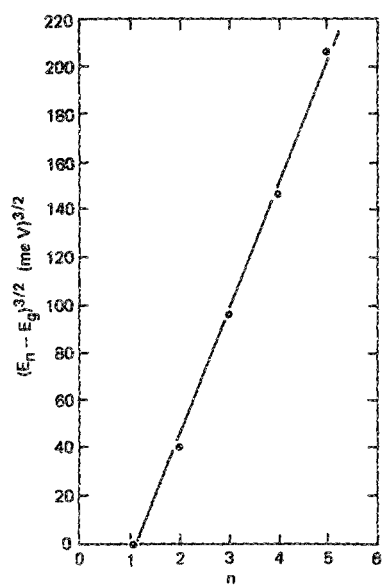


FIG. 7. Extrema energy to the three-half power vs the point number $n$, where $E_g=E_1$.

mined by the sample itself. Our studies show that PR is a very sensitive technique. We observed PR signals using pump beam intensities as low as that of the probe (reflected) beam ($\sim 1\ \mu W/cm^2$). The sample quality (defect, background impurity, etc.), the surface condition, the inhomogeneous field, and the fluctuation of the AlAs mole fractions should affect the PR line shape. From our results, we believe that the line shape having more numbers of oscillations is indicative of smaller broadening and more uniformity of the samples. A semiquantitative relationship between the PR line shape and the sample quality is thus possible, but further work is needed.

Second, the surface field homogeneity can be estimated for samples which show oscillations. We have calculated the surface field for sample 1583 (which should be averaged over the pump beam penetration distance) to be 10.3 kV/cm. Assuming that the Fermi level is pinned at the midgap on the surface and the background impurity is $5\times10^{15}$ $cm^{-3}$, then the band bending near the surface is 0.73 eV and the depletion region is estimated to be 1.1 $\mu$m. The pump beam penetration depth is about 0.5 $\mu$m since the absorption coefficient of $Al_xGa_{1-x}As$[14] ($x\leqslant 0.45$) at the modulation wavelength 6328 Å is about $2\times10^4$ $cm^{-1}$. Therefore, in the modulation-dominated region, the field spatial inhomogeneity is obvious. However, Grover *et al.*[35] demonstrated that the field homogeneity has a lesser effect than the broadening on the electroreflectance line shape. There are still many oscillations even for high-field inhomogeneity.

Third, the more complicated form of PR line shapes in $Al_xGa_{1-x}As$ compared to that in quantum wells is partially caused by the relatively strong impurity-related transitions, and the energy degeneracy of the heavy and light holes at the Brillouin zone center in bulk samples (if the splitting of the heavy and light holes is not assumed). Pond and Handler[36] observed the interference between the light and heavy-hole contributions to the electroreflectance signal which is predicted by the modulation reflectance theory. In such a spectra, the heavy- and light-hole signals should have different intensities and oscillation periods due to their different effective masses. According to the theory, the period is determined by the reduced mass of the electron and hole pair rather than the hole mass alone. Thus, the difference between the two reduced masses (heavy and light hole) is small, $\mu_{hh}/\mu_{lh} = 1.5$. In addition, the light-hole transition probability should be smaller than that of the heavy hole due to the small density of states. These reasons make it difficult to distinguish the light-hole signal from that of the dominant heavy hole. At this point, we are not sure how important the heavy- and light-hole signal interference is in determining the PR line shape. However, as we indicated previously, the impurity-related transitions really play an important role. In fact, the impurity line usually extends to and mixes with the dominant intrinsic signal and changes the total line shape drastically as shown in Fig. 4. We were able to distinguish these two signals, and a more detailed analysis will be presented elsewhere.

Fourth, some samples show low-field-like PR line shapes and the spectrum in Fig. 3 is one of them. Although several other mechanisms have been proposed to account for the PR spectrum in addition to the surface field modulation, like band filling, heating, and carrier screening, the experimental studies by Shay,[3] Aspnes,[37] and Shanabrook *et al.*[13] demonstrated that the surface field modulation should be the dominant one at moderate light intensities. Hence, the use of the Aspnes TDFF to fitting the low-field-like line shapes should be reasonable. Several factors lead to such a line shape: a low surface field, large broadening, inhomogeneous field, and fluctuations in the AlAs mole fraction. Since there is no reason to say that the surface fields in these samples are lower than those showing oscillations, and since the field inhomogeneities have a lesser effect on the line shape as stated above, we believe that the dominant one is the fluctuation of the AlAs mole fraction along the growth direction. This conclusion is strongly supported by the nuclear profiling results. As one can clearly see, sample 1583 is more uniform than sample 1841. This result also supports our claim that more oscillations in PR spectra correspond to better alloy quality.

Finally, the most important remaining problem is the fitting of these different line shapes to a universal function by an appropriate choice of parameters related to the material itself, such as the AlAs mole fraction, broadening, surface band bending, and impurity concentration, etc. We believe that such a fitting is possible and will be very useful in the material characterization. To do so, one needs a deeper understanding of the nature of PR. A combination of PR and electroreflectance (via a Schottky barrier) can be very helpful in this regard. Other mechanisms like the band filling (phase space filling), electron screening, and the lifting of degeneracy on PR may become more important under high pump intensity, at low temperature, and are still poorly understood and should be studied further. We believe that the PR results from the $Al_xGa_{1-x}As$ system can be used to characterize other bulk materials.

## IV. SUMMARY AND CONCLUSIONS

The PR spectra of the bulk $Al_xGa_{1-x}As$ alloys with $x\leqslant 0.45$ have been studied. The line shapes observed in the different samples indicate that PR is very sensitive to the material quality, the surface condition, and to the impurity background. The energy gaps derived from the PR spectra are compared with those from the absorption spectra, and a good agreement is demonstrated. The relationship between the energy gaps and the AlAs mole fraction $x$ was established with the $x$ values determined directly from the nuclear resonant reaction analysis (NRRA) profiling technique. Franz–Keldysh oscillations were observed in the PR spectra of several samples, and from the period of oscillations we calculated the electric field near the semiconductor surface. From our analysis we believe that the increased number of oscillations in PR spectra corresponds to better sample quality, in general. By comparing PR line shapes and the NRRA profiling results, and by estimating the surface field homogeneity, we conclude that the low-field-like line shape is mainly caused by the fluctuation of the Al distribution in the alloys along the growth direction. The low-energy features were assigned to be the impurity-related transitions, but the importance of light-hole-related transitions in the PR line

shape is yet to be established. All of the above results and detailed discussions demonstrate that PR can be an extremely convenient and useful technique in material characterization. It is also helpful in understanding the many-body interactions via the photoexcited carriers.

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